THE ORIGIN OF PRESOLAR SILICA GRAINS IN AGB STARS. M. Bose[1], C. Floss[1], F. J. Stadermann[1], R. M. Stroud[2], and A. K. Speck[3]. [1]Laboratory for Space Sciences and Physics Department, Washington University, St. Louis MO, 63130, USA. [2]Naval Research Laboratory, Washington DC, 20375, USA. [3]University of Missouri, Columbia MO, 65211, USA. (Email: mbose@physics.wustl.edu)

Introduction: Oxide and silicate grains have been identified by infrared (IR) spectroscopy to be a major component of the dust shells in O-rich Asymptotic Giant Branch (AGB) stars. Recently, such grains have also been identified in the laboratory through investigations of meteoritic presolar grains. Coordinated isotopic and elemental measurements of these presolar grains can enhance our understanding of dust formation processes in O-rich environments and also aid in the interpretation of IR spectra.

The IR spectra of about half of all AGB stars exhibit a 13 μm feature [1]. A variety of phases have been proposed as its carrier including SiO_2 whose extinction spectra similarly show a 13 μm peak [2]. Here we report on two silica grains identified in the primitive carbonaceous chondrite ALHA77307 [3], and explore the possibility that silica may be the carrier of the 13 μm feature associated with evolved stars.

Experimental Methods: Phases with anomalous O-isotopic compositions were identified in a polished thin-section of ALHA77307 with the NanoSIMS 50. A primary Cs^+ beam of less than 1 pA was rastered over $10{\times}10~\mu m^2$ matrix areas. Secondary electrons as well as $^{12,13}C^-$ and $^{16,17,18}O^-$ secondary ions were collected in multi-collection mode, with total integration times of ~2 hours per area. Following the isotopic measurements, the elemental compositions of the O-anomalous grains were measured with the PHI 700 Scanning Auger Nanoprobe at electron beam settings of 10 keV and 0.25 nA. Elemental maps were acquired for selected grains.

After the Auger Nanoprobe measurements, one of the silica grains found was measured by Transmission Electron Microscopy (TEM). The TEM sample was prepared by focused ion beam (FIB) extraction using the Nova NanoLab 600, and was then characterized in a 200 kV JEOL 2200FS TEM at the Naval Research Laboratory. Finally, an additional O-isotopic measurement of the extracted FIB section was done with the NanoSIMS at Washington University.

Results: Our isotopic survey of ALHA77307 led to the identification of sixty-six presolar grains, two of which were later identified as likely silica grains. Figure 1 shows the O-isotopic compositions of these two silica grains as well as two similar grains identified in other studies [4, 5]. All four of the silica grains found to date are ^{17}O-rich, with near-solar $^{18}O/^{16}O$ ratios, and therefore can be classified as group 1 grains [6]. Outflows of low-mass AGB stars that have undergone first dredge-up are likely sources for group

1 grains. The identification as possible silica is based on Auger Nanoprobe data, which show that both grains have compositions consistent with SiO_2 (O/Si = 2.2±0.3; Figure 2); Mg and Fe contents in the grains are below detection limits (< 3 at.%).

Grain 21420 was extracted using the FIB technique and studied in the TEM. The measurements showed the presence of a $50{\times}140~nm^2$ Si-rich area, adjacent to a 300 nm Mg- and Fe-bearing grain. NanoSIMS measurements on the extracted slice showed a solar O-isotopic composition for the Mg- and Fe-bearing grain, but the size of the Si-rich region was insufficient for definitive structural or isotopic characterization.

Three of the silica grains identified to date come from ALHA77307. Based on the data from this study, the abundance of presolar silica is 4 ppm and makes up ~4% of all presolar silicates found in this meteorite.
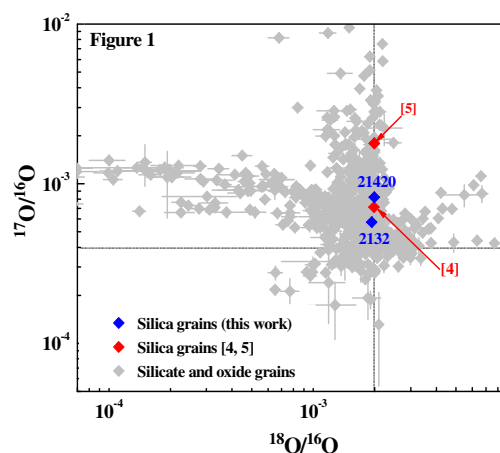
Figure 1. *Oxygen three isotope plot showing silica grains in ALHA77307 [this work, 4] and QUE 99177 [5]. Comparison data (light gray) are from http://presolar.wustl.edu/~pgd/.*

Formation Conditions of Silica Grains: Presolar silica has been described as "mythical" because equilibrium condensation calculations show that Si forms Mg-rich silicates (and not SiO_2) in circumstellar shells [7]. However, silica may originate during rapid cooling under non-equilibrium conditions in stars with low Mg/Si ratios [8]. Evidence pointing towards non-equilibrium condensation of amorphous silica aggregates also comes from laboratory experiments [9], and models of kinetically condensing systems [10]. The latter study suggests that SiO_2 may form in circumstellar outflows because condensation of

metallic Fe onto forsterite prevents the formation of enstatite. Thus, the silica stardust grains found in this and other studies may have formed through kinetically driven reactions in stellar environments.
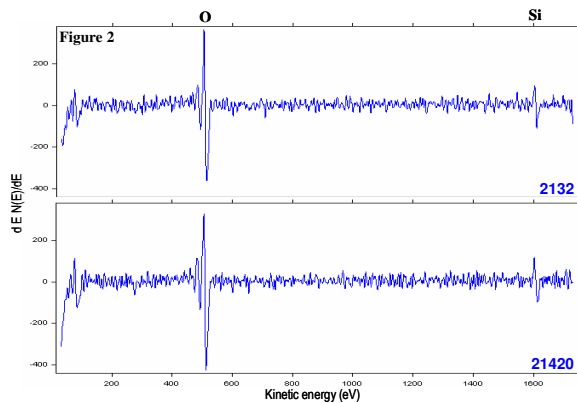
Figure 2: *Differentiated Auger elemental spectra of the silica grains.*

Astronomical Implications: Because of the difficulties in modeling the condensation of silica, as discussed above, silica has frequently been discounted as a possible carrier of the 13 µm spectral feature. However, with the laboratory observation of silica-like phases in the presolar grain inventory, it is worthwhile revisiting this question.

As noted above, the 13 μm feature is observed in about half of all AGB stars; among these it is predominantly associated with semi-regular (SR) variable stars and, to a lesser extent, is found in Mira variable stars [e.g., 1]. A host of minerals including corundum, spinel, Ti-oxide, and core-mantle-grains composed of, e.g., Al_2O_3 cores and silicate mantles, or TiO_2 cores and corundum mantles, have been suggested as possible carriers of the 13 μm feature [11–15]. Various models have attempted to constrain the carrier phase of the 13 μm feature, by incorporating parameters such as grain composition, grain shape and size, as well as a host of other factors, including varying mass-loss rates. Spinel is one leading candidate as carrier of the 13 µm feature, but it exhibits secondary features at other wavelengths, which are not observed in stellar spectra [16]. Titanium-oxide grains provide a good match as well, but the low abundance of Ti in stars reduces the likely contribution of this phase to the dust emission at 13 µm [e.g., 14]. Grains with core-mantle structures can also be matched to the 13 µm feature, but only under limited conditions that make their viability as carriers of this feature rather unlikely [11]. Corundum (α-Al_2O_3) remains a viable candidate as well, with only small amounts (~1%) needed to produce the 13 µm feature [17].

Finally, it has been suggested that the 13 µm feature may be associated with SiO_2, or other highly polymerized silicates [2]. Laboratory extinction spectra of the three main polytypes of crystalline SiO_2, quartz, tridymite and cristobalite, as well as amorphous silicon dioxide and silica glass exhibit the 13 µm feature [2, 18], although detailed modeling remains problematic [18]. However, arguments have also been raised against SiO_2 as the carrier of the 13 µm feature, including opacity issues leading to reduced emission at the IR wavelengths, and the absence of a correlation between the 13 µm and 10 µm silicate features in all stellar spectra [e.g., 13]. Nevertheless, if silica is indeed a product of non-equilibrium condensation, it may be a feasible carrier of this spectral feature in SR variable stars. These stars have lower mass loss rates than Mira variable stars, leading to a lower density at the dust-forming radius [19]. Non-equilibrium conditions prevail at these lower densities, with the result that silica is less likely to react with other circumstellar species (e.g., MgO) to form magnesian silicates and, thus, may survive as SiO_2.

In summary, although the carrier of the 13 µm spectral feature remains uncertain (and, indeed, may require more than one phase), presolar silica grains, like those reported here, could be one plausible contributor. Further modeling of laboratory analogues of different crystalline and amorphous forms of SiO_2 should help evaluate this possibility. Determination of the crystal structure of the presolar silica grains found in primitive meteorites can provide additional constraints. We plan to study additional silica grains with coordinated NanoSIMS-Auger-TEM in the future.

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